

# POLYCYCLIC AROMATIC COMPOUNDS IN FLUIDIZED BED COMBUSTION OF COAL

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## Introduction

Polycyclic aromatic compounds (PAC) have been determined in the gas and particulate effluents from fluidized bed combustion of coal, lignite, and oil shale by a number of investigators. A bibliography of the reports of these investigations is contained in a paper by Chiu, Walsh, Beer, and Eiemann (1983). The concentrations of PAC in the effluents are often quite low, but depend upon the combustor configuration and operating conditions. The goal of the present investigation is to develop a mechanism consistent with measured rates of formation and destruction of PAC in atmospheric pressure fluidized bed combustion (AFBC), so that a rational approach may be taken to adjustment of conditions for minimization of PAC in the effluents. PAC are determined in samples taken from the freeboard (space above the bed), rather than from the exhaust, to observe the evolution of the PAC distribution within the combustor. Mass fractions of the most abundant PAC observed in the freeboard during AFBC of high volatile bituminous coal were reported by Chiu, et al (1983). Some correlation of the rates of disappearance of PAC with particle concentration was noted by Dutta, Chiu, Walsh, Beer, and Eiemann (1983). In the present paper theoretical estimates of the rates at which PAC might be consumed by heterogeneous reactions are compared with experimental rates estimated from PAC profiles determined by Chiu et al (1983).

## Experimental

The PAC were determined in samples taken in the MIT AFBC Research Facility. A detailed description of the facility may be found in a report by Beér, Sarofim, Sandhu, Andrei, Bachovchin, Chan, Chaung, and Sprouse (1981). The combustor has a square cross section 0.6 m x 0.6 m and a height, from the combustion air distributor to the outlet, of 4.4 m. Previous publications describe the procedures used for sampling, separation, and identification of PAC (Chiu et al, 1983), permanent gases (Walsh, Chaung, Dutta, Beér, and Sarofim, 1982b), and bed particles (Walsh et al, 1982a; 1982b).

The AFBC was fired with Kentucky No. 9 high volatile bituminous coal crushed to minus 6.35 mm. The apparent ASTM rank of this coal was high volatile B bituminous. Other coal properties are given by Chiu et al (1983). The bed material was Reed limestone (middle ledge) crushed to minus 6.35 mm.

The experimental conditions at which the PAC were measured are listed in Table 1. In the specification of bed heights, sampling points, etc., the height above the distributor is designated by the symbol  $z$ . The top of the fluidized bed is at  $z = L_f$ . Axial profiles of temperature and the mole fractions of  $O_2$ ,  $CO$ ,  $NO$ , and  $CH_4$  are shown in Figure 1. The freeboard temperature in this run was relatively low, due to cooling of the combustion products by heat exchanger tubes

Table 1. Experimental Data

|   |                           |
|---|---------------------------|
| Run Number  | K10                       |
| Fraction of theoretical air:  | 1.01                      |
| Superficial gas velocity based on average bed temperature and pressure at $z = 0$ : | 1.20 m/s                  |
| Minimum fluidization velocity:  | 0.18 m/s                  |
| Fluidized bed height:   | 0.65 m                    |
| Calcium/sulfur feed ratio:  | 3.7 kmol/kmol             |
| Solid density of bed char particles:  | 336 kg/m <sup>3</sup>     |
| Solid density of bed stone particles:   | 2163 kg/m <sup>3</sup>    |
| Freeboard pressure:   | 102.6 kPa                 |
| Average freeboard temperature from $z = 2.0$ to $3.8$ m:                            | 964 K                     |
| Average freeboard gas velocity from $z = 2.0$ to $3.8$ m:                           | 1.18 m/s                  |
| Mass-based geometric mean size of elutriated char particles:                        | 72 $\mu$ m                |
| Geometric standard deviation of elutriated char particle size distribution:         | 1.82                      |
| Specific surface area-based geometric mean size of elutriated char particles:       | 60 $\mu$ m                |
| Mass-based geometric mean size of elutriated stone particles:                       | 74 $\mu$ m                |
| Geometric standard deviation of elutriated stone particle size distribution:        | 2.67                      |
| Specific surface area-based geometric mean size of elutriated stone particles:      | 46 $\mu$ m                |
| Terminal velocity of mean size elutriated char particles:                           | 0.012 m/s                 |
| Terminal velocity of mean size elutriated stone particles:                          | 0.046 m/s                 |
| Sherwood numbers of elutriated char and stone particles                             | $\sim 2$                  |
| Total flowrate of elutriate:  | 0.01167 kg/s              |
| Mass fraction char in elutriate:  | 0.063                     |
| Concentration of entrained char in the freeboard:                                   | 0.00169 kg/m <sup>3</sup> |
| Concentration of entrained stone in the freeboard:                                  | 0.0260 kg/m <sup>3</sup>  |

located at  $z = 0.8, 1.0, \text{ and } 1.2 \text{ m}$ . The bed material at the start of the run was fresh calcined limestone, therefore the ratio of  $\text{CaO}$  to  $\text{CaSO}_4$  in the bed is much higher than would be present after sufficient time had elapsed to achieve steady bed composition.

The mole fractions of some of the PAC identified in samples taken from the freeboard at  $z = 1.6, 2.1 \text{ and } 3.8 \text{ m}$  are given in Table 2. These mole fractions are based on the volume of dry gas sampled. Table 2 includes all of the compounds which were identified at more than one height. The PAC mole fractions are plotted vs height above the distributor in Figure 2. Most of the profiles indicate only net destruction of PAC in the region of the measurements. There is some evidence that production of PAC is occurring in the freeboard, and a hint that there may be a peak mole fraction resulting from the combined production and consumption processes which tends to shift to longer times (heights) with increasing molecular weight (compare the profiles of naphthalene, phenanthrene, anthracene, and chrysene in Figures 2a and 2g). However, much significance cannot be attached to changes in the slopes of the profiles, because the uncertainty in the reported mole fractions is approximately  $\pm 100, -50\%$ . Pseudo-first order rate coefficients,  $k_{\text{exp}}$ , were derived from the mole fractions at  $z = 2.1 \text{ and } 3.8 \text{ m}$ :

$$u_o \frac{dC_{\text{PAC}}}{dz} = -k_{\text{exp}} C_{\text{PAC}} \quad (1)$$

$$k_{\text{exp}} = - \frac{u_o}{z_2 - z_1} \ln \frac{X_{\text{PAC},2}}{X_{\text{PAC},1}} \quad T, P, \Sigma C_j \approx \text{constant} \quad (2)$$

The definitions of symbols may be found in the Nomenclature. The values of  $k_{\text{exp}}$  for the PAC identified at  $z = 2.1 \text{ and } 3.8 \text{ m}$  are given in Table 2. Lack of a strong dependence of the experimental rate coefficients on oxygen, temperature, or compound, and their tendency to increase with increasing particle concentration (Dutta et al, 1983), suggested that the PAC destruction process might be heterogeneous, with significant resistance from extraparticle diffusion.

#### Heterogeneous Mechanism for Destruction of PAC

The mechanism of the transformations of PAC in the freeboard gas/particle mixture is undoubtedly extremely complicated. The investigations of Bittner and Howard (1981a; 1981b) and Howard and Bittner (1983) on the reactions of hydrocarbon species in benzene/oxygen flames give an indication of the magnitude of the problems which may be encountered in the freeboard gas/particle mixture. Suppose that we neglect production of a given PAC by char devolatilization, synthesis from lighter species, or formation by degradation of heavier species; and that each PAC is degraded to non-PAC in irreversible, heterogeneous reactions with solid species  $S_i$ :



We assume that the freeboard gas/particle mixture is in steady plug flow, and neglect axial dispersion. A rate expression based on this mechanism is:

Table 2. Polycyclic Aromatic Compounds Identified at Two or Three Heights in the Freeboard

| Compound                   | Mole Fractions $\times 10^9$<br>(dry basis) |                   |                   | $k_{exp}$<br>(Hz) | $(\epsilon v)_{PAC}^b$<br>( $m^3/kmol$ ) | $D_{PAC-N_2}$<br>$\times 10^{42}$<br>( $m^2/s$ ) |
|----------------------------|---|-------------------|-------------------|-------------------|--|--|
|                            | $z = 1.6m$                                  | $2.1m$            | $3.8m$            |                   |  |  |
| benzofuran                 | 22.5  | 5.72 <sup>a</sup> | .093 <sup>a</sup> | 2.9               | .109                                     | .577   |
| naphthalene                | 991   | 324               | 1.40              | 3.8               | .140                                     | .515   |
| benzo(b)thiophene          | 112   | 33.0              | .070              | 4.3               | .120                                     | .548   |
| quinoline                  | 17.1  | 2.11              | -                 | -                 | -  | -  |
| 2-methylnaphthalene        | 25.6  | 8.81              | .244              | 2.5               | .161                                     | .479   |
| 3-methylbenzo(b)thiophene  | 2.80  | .10               | -                 | -                 | -  | -  |
| 1-methylnaphthalene        | 25.0  | 6.23              | .04 <sup>a</sup>  | 3.5               | .161                                     | .479   |
| biphenyl                   | 88.1  | 25.7              | .182              | 3.4               | .177                                     | .455   |
| ethylnaphthalene           | 2.95  | .90               | -                 | -                 | -  | -  |
| acenaphthylene             | 45.4  | 11.2              | -                 | -                 | -  | -  |
| acenaphthene               | 2.19  | .41               | -                 | -                 | -  | -  |
| 1-cyanonaphthalene         | 6.53  | 1.15              | -                 | -                 | -  | -  |
| dibenzofuran               | 44.3  | 19.8              | .108              | 3.6               | .159                                     | .475   |
| methyldibenzofuran         | .79   | .59               | -                 | -                 | -  | -  |
| 9-fluorenone               | 10.3  | 2.50              | .109              | 2.2               | .195                                     | .431   |
| dibenzothiophene           | 23.8  | 9.49              | .193              | 2.7               | .170                                     | .458   |
| phenanthrene               | 125   | 64.7              | .204              | 4.0               | .190                                     | .436   |
| anthracene                 | 9.53  | 8.43              | -                 | -                 | -  | -  |
| fluoranthene               | 6.07  | 14.6              | -                 | -                 | -  | -  |
| benzo(def)dibenzothiophene | 2.43  | 7.34              | -                 | -                 | -  | -  |
| pyrene                     | 4.13  | 12.0              | -                 | -                 | -  | -  |
| chrysene                   | 1.09  | 3.42              | .101              | 2.4               | .240                                     | .385   |

a. In the gas chromatographic separation this compound coeluted with another compound. The amount indicated is an estimate based on the relative intensities of peaks characteristic of the two compounds in the mass spectrum.

b. Fuller, Schettler, and Giddings (1966).

$$u_o \frac{dC_{PAC}}{dz} = - \sum k'_{PAC,i} C_{PAC} \quad 3)$$

where  $k'_{PAC,i}$ , the effective rate coefficient for the heterogeneous reaction of  $PAC$  with solid species  $i$ , contains the sequential processes of diffusion of  $PAC$  to the surfaces of particles, and the heterogeneous reaction at the surfaces:

$$k'_{PAC,i} = \frac{\rho_i}{\frac{6Sh_i D_{PAC}}{\phi_i \rho_i^* d_i^2} + k_{PAC,i} S_i \eta_{PAC,i}} \quad 4)$$

In order to evaluate this rate coefficient we require estimates of the concentration, diameter, solid density, and sphericity of each particle species, and the diffusion coefficients of the  $PAC$ .

#### Freeboard Particle Properties

High particle concentration is an important feature of the freeboard flow, however precise measurements of the particle concentrations are difficult. The problems arise from the facts that large particles ( $> 300 \mu m$ ) are moving both upward and downward with distributed velocities; and that small particles ( $\leq 300 \mu m$ ), which are moving primarily upward, have velocities which depend on particle size and density. Isokinetic sampling of the particles is therefore not possible. Concentrations of particles in the bed are usually determined from measurements of differential pressure. This technique can only be used in the freeboard at low superficial velocity when pressure fluctuations are not large with respect to the changes in pressure due to the presence of particles. This condition is usually not met during combustion measurements. An alternative, approximate method for the determination of particle concentrations has been developed in order to estimate the rates of heterogeneous reactions.

The particles are classified by their terminal velocities into two groups: large particles (terminal velocities greater than the mean gas velocity) and small particles (terminal velocity less than the mean gas velocity). The dependence of the flux of large particles on height in the freeboard is measured by catching descending particles in cups placed at several heights above the bed (Walsh, Mayo, and Beér, 1983). The concentration of large particles is estimated from the flux measurements by a method proposed by Walsh, Yokozeki, and Beér (1982). The basis of this estimate is an assumed initial velocity distribution (Gaussian) of particles leaving the bed surface. For a given particle flux at the bed surface, a given standard deviation of the velocity distribution, and the assumptions of negligible drag and total reflux; both the flux and concentration of particles at a given height are uniquely determined. In this way the large particle concentrations can be estimated from the flux measurements.

The small entrained particles are precipitated by cyclones from the exhaust, so their flux at the freeboard exit is known. By assuming that they move only upward at a constant velocity equal to the mean gas velocity minus the terminal velocity of the mean size particles, their concentration can also be estimated. Errors in this estimate may arise from:

1. Cyclone collection efficiency less than 1.
2.  $\text{SO}_2$  and/or  $\text{CO}_2$  reactions with stone and combustion of char in the freeboard<sup>2</sup> and exhaust pipe before the collection point.
3. Refluxing of the small particles down the freeboard wall (Pemberton and Davidson, 1983).

Probably the most important problem, however, is in the initial assumption: that the particles can be divided into two groups having mean sizes independent of height. The particle size and velocity distributions undergo continuous evolution from the top of the bed (and even within the bed itself) to the combustor outlet. A more detailed calculation can be made by machine computation, but the value of such an effort is to some extent negated by the lack of experimental data with which to compare the predictions.

The mass-based particle size distributions of the stone and char particles in a sample of the elutriate precipitated in the cyclones during the present experiment are shown in Figure 3. The mass fractions of stone and char in the elutriate were calculated from the heating value of the material in each size range. The heating value of pure char was estimated using Eulong's formula with a composition derived from the ultimate and proximate analyses of the coal. The specific surface area-based mean particle sizes were calculated from the standard deviation and mass-based mean sizes (Irani and Callis, 1963). The sphericity of the entrained stone and char particles was estimated to be 0.5; their solid densities were assumed to be the same as those of the larger stone and char particles taken from the bed. Terminal velocities of the particles were calculated using the correlation of Pettyjohn and Christiansen (1948). The properties of the entrained particles are summarized in Table 1.

The refluxing particles were assumed to have the same size, sphericity, and solid density as bed particles. Specific surface area-based mean sizes of the bed stone and char particles were 780 and 2900  $\mu\text{m}$ , respectively. The mass fraction of char in the bed and refluxing particles is only 0.43 wt %.

Total particle concentrations estimated in the present experiment by the approximate methods outlined above are shown in Figure 4. The bed particle concentrations were determined from measurements of differential pressure, the refluxing particles from the samples collected in the cups, and the entrained particles from the cyclone catch. An explanation of the second curve on Figure 4 is given in the Discussion section.

#### Molecular Diffusion Coefficients of Polycyclic Aromatic Compounds

Experimental measurements of the molecular diffusion coefficients of PAC are rare. Diffusion coefficients in air at atmospheric pressure have been reported for naphthalene and biphenyl at 298 K, anthracene at 372 K (Mack, 1925), and biphenyl at 491 K (Gilliland, 1934). Howard and Bittner (1983) determined diffusion coefficients of heavy species ( $W = 200$  to  $750$  kg/kmol) from concentration profiles measured in a near-sooting benzene/oxygen/argon flame (equivalence ratio 1.8), in a region of the flame where the slopes of the species profiles,  $d\ln C/dz$ , were determined only by diffusion. The pressure in the flame, and the temperature in the region of the measurements were 2.67 kPa and 1800 K, respectively. Assuming that the species observed were polycyclic aromatic hydrocarbons, Howard and Bittner estimated their diffusion coefficients at 300 K from the correlation of Fuller, Schettler, and Giddings (1966), using the temperature dependence

derived from the Lennard-Jones 12-6 potential, as recommended by Sherwood, Pigford, and Wilke (1975). The approximation given by Fristrom and Westenberg (1965) to the temperature dependence of the diffusion collision integral was used. The correlation of Fuller et al, with this modification, becomes (in SI units):

$$D_{AB} = \frac{1.60 \times 10^{-4} T^{1.67} \left( \frac{1}{W_A} + \frac{1}{W_B} \right)^{1/2}}{P \left[ (\Sigma v)_A^{1/3} + (\Sigma v)_B^{1/3} \right]^2} \quad \text{m}^2/\text{s} \quad (5)$$

Howard and Bittner (1983) reported good agreement between their experimental diffusion coefficients and those estimated from the modified correlation. The discrepancies between estimated values and the experimental values for PAC reported by Mack (1925), Gilliland (1934), and Howard and Bittner (1983) are  $\pm 10$  to 15%.

Estimates of the binary diffusion coefficients of PAC in  $N_2$  using Equation 5 are listed in Table 2. The procedure given by Fuller et al (1966) for the estimation of the molecular diffusion volumes from atomic diffusion volumes was followed. The volume increment  $-0.0202 \text{ m}^3/\text{kmol}$  was added for each aromatic and heterocyclic ring, but not for the ring containing the keto group in 9-fluorenone.

### Discussion

An upper limit on the rate coefficient for the surface reaction can be estimated from the product of the frequency of collisions of the PAC with unit area of a surface and the particle external surface area per unit volume of gas:

$$k_{PAC,i} S_{i,PAC,i} \rho_i \leq \left( \frac{RT}{2\pi W_{PAC}} \right)^{1/2} \frac{6}{\phi_i \rho_i d_i} \rho_i \quad (6)$$

Using the entrained particle properties in Table 1 the upper limits for naphthalene at 964 K are 310 and 100 Hz for the stone and char particles, respectively.

The rate coefficient for the diffusion process is equal to

$$\frac{6Sh_i D_{PAC}}{\phi_i \rho_i d_i^2} \rho_i \quad (7)$$

Using the elutriated particle properties from Table 1 and the diffusion coefficient for naphthalene in  $N_2$  from Table 2, values of 7 and 2 Hz are obtained for the stone and char, respectively. The rate coefficient for the surface reaction is therefore estimated to be more rapid than that for external diffusion for values of the sticking coefficients greater than about 0.02. If the sticking coefficient were sufficiently large, say  $\geq 0.1$ , the experimental rate coefficient would be approximately equal to the diffusion limited rate coefficient given by Equation 7.

In general the concentration of particles,  $\rho_i$ , is a function of height, as shown in Figure 4. However, the quantity  $6Sh_i \rho_i / \phi_i \rho_i d_i^2$  is dominated by the approximately constant concentration of small particles over most of the height of the freeboard under the present conditions, shown by the second curve in Figure 4. We may therefore base estimates of the diffusion limited rate coefficients from Equation 7

on the properties of the small particles alone.

The calculated rate coefficients for destruction of PAC by diffusion limited surface reactions on the small stone and char particles are plotted vs. PAC diffusion coefficient in Figure 5. The rate coefficient for simultaneous diffusion controlled destruction on both types of particles is the sum of the values shown for stone and char. The data points are the experimental rate coefficients determined from the PAC mole fractions at  $z = 2.1$  and  $3.8$  m.

Any values of the experimental rate coefficients which are equal to, or less than the theoretical upper limit (sum of stone and char values) are consistent with the model. Since this condition is not difficult to satisfy the location of the experimental points below the upper limit is little more than a justification for further testing and examination of the model. However, the consistency of the data points (factor of 2) is interesting. If the similarity of the values of the experimental rate coefficients is not due to significant diffusion resistance, then the rate coefficients for the net destruction of PAC by heterogeneous and/or homogeneous reactions are rather insensitive to molecular structure. The location of the experimental points between the rate coefficients for diffusion limited reaction on stone and char is an indication that at least part of the stone is participating in the reaction. Such a conclusion is not justified however, considering the assumptions and approximations used in the model and calculations. The reaction might be occurring only on the char. If the PAC destruction were only via heterogeneous reactions the intercept of a line through the data points would be zero at  $D_{PAC} = 0$ . The extrapolated value is approximately  $0.7$  Hz.

### Conclusion

A heterogeneous mechanism for destruction of PAC in the AFBC freeboard could not be ruled out by comparison with the rates of disappearance of 2 to 4 ring PAC at one set of operating conditions. If heterogeneous reactions are, in fact, responsible for PAC destruction, a more detailed model accounting for production of PAC and finite rates of the surface reactions may be needed. The relative importance of stone and char particles can only be determined by additional measurements over a range of ratios of the char/stone concentrations. The possibility that only a fraction of the surface area of each type of particle is utilized, due to nonuniform surface composition, will be difficult to assess. Better spatial resolution of the PAC profiles is needed.

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### Nomenclature

|                |  |
|----------------|--|
| $C_j$          | concentration of gaseous species $j$ , kmol/m <sup>3</sup>   |
| $d_i$          | specific surface area-based geometric mean diameter of particles $i$ , m   |
| $D_{AB}$       | binary diffusion coefficient, m <sup>2</sup> /s  |
| $D_{PAC}$      | diffusion coefficient of PAC in the freeboard gas, assumed to be equal to $D_{PAC-N_2}$ , m <sup>2</sup> /s      |
| $k_{exp}$      | experimental first order rate coefficient, Hz  |
| $k_{j,i}$      | rate coefficient for surface reaction of gaseous species $j$ with particles $i$ , m/s                            |
| $k'_{j,i}$     | effective first order rate coefficient for heterogeneous reaction of gaseous species $j$ with particles $i$ , Hz |
| $L_f$          | fluidized bed height, m  |
| $P$            | pressure, Pa   |
| $R$            | gas constant = 8314 J/kmol.K   |
| $S_i$          | BET specific surface area of particles $i$ , m <sup>2</sup> /kg  |
| $Sh_i$         | Sherwood number for particles $i$ , dimensionless  |
| $T$            | temperature, K   |
| $u_o$          | superficial (empty tube) gas velocity, m/s   |
| $W_j$          | molecular weight of species $j$ , kg/kmol  |
| $X_j$          | mole fraction of gaseous species $j$ , dimensionless   |
| $z$            | height above air distributor, m  |
| $\eta_{j,i}$   | effectiveness factor for reaction of gaseous species $j$ with particles $i$ , dimensionless                      |
| $\rho_i$       | concentration of particles $i$ , kg/m <sup>3</sup>   |
| $\rho_i^*$     | solid density of particles $i$ , kg/m <sup>3</sup>   |
| $\phi_i$       | sphericity of particles $i$ , dimensionless  |
| $(\Sigma v)_j$ | sum of atomic diffusion volumes for molecules of gaseous species $j$ , m <sup>3</sup> /kmol                      |

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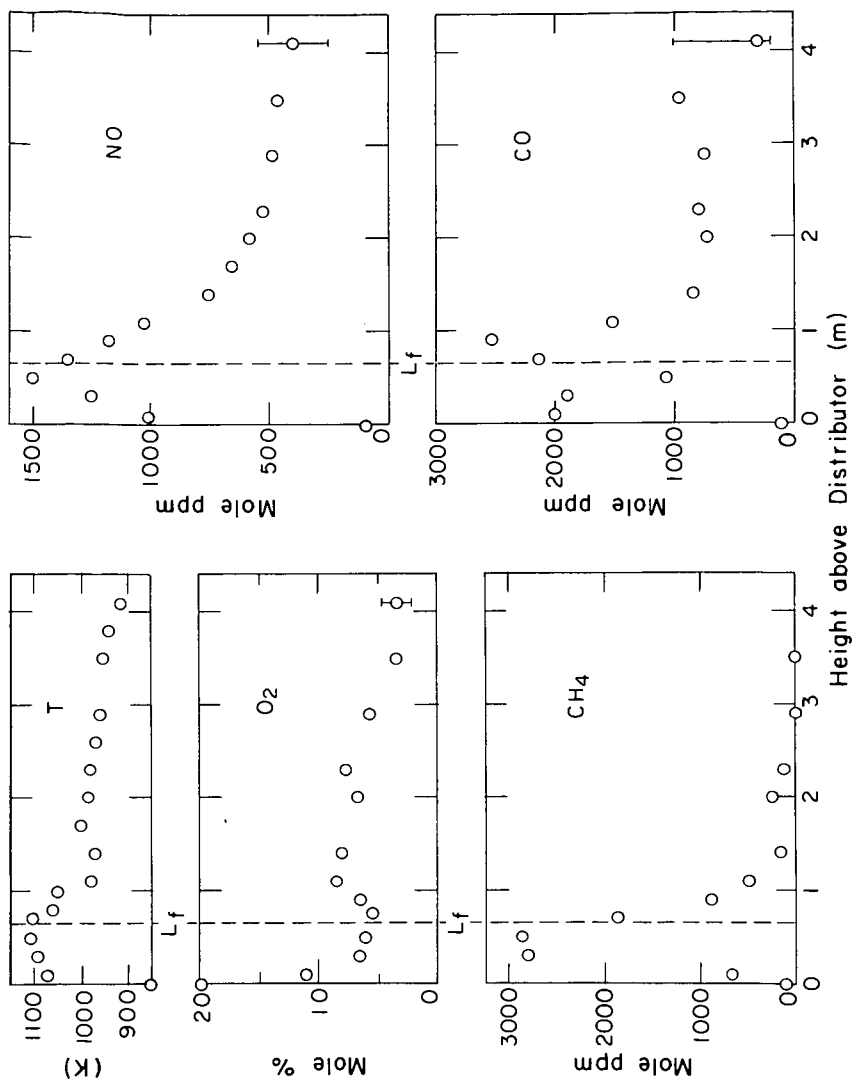


Figure 1. Temperature and mole fractions of light gaseous species (dry basis) vs. height in the bed and freeboard.

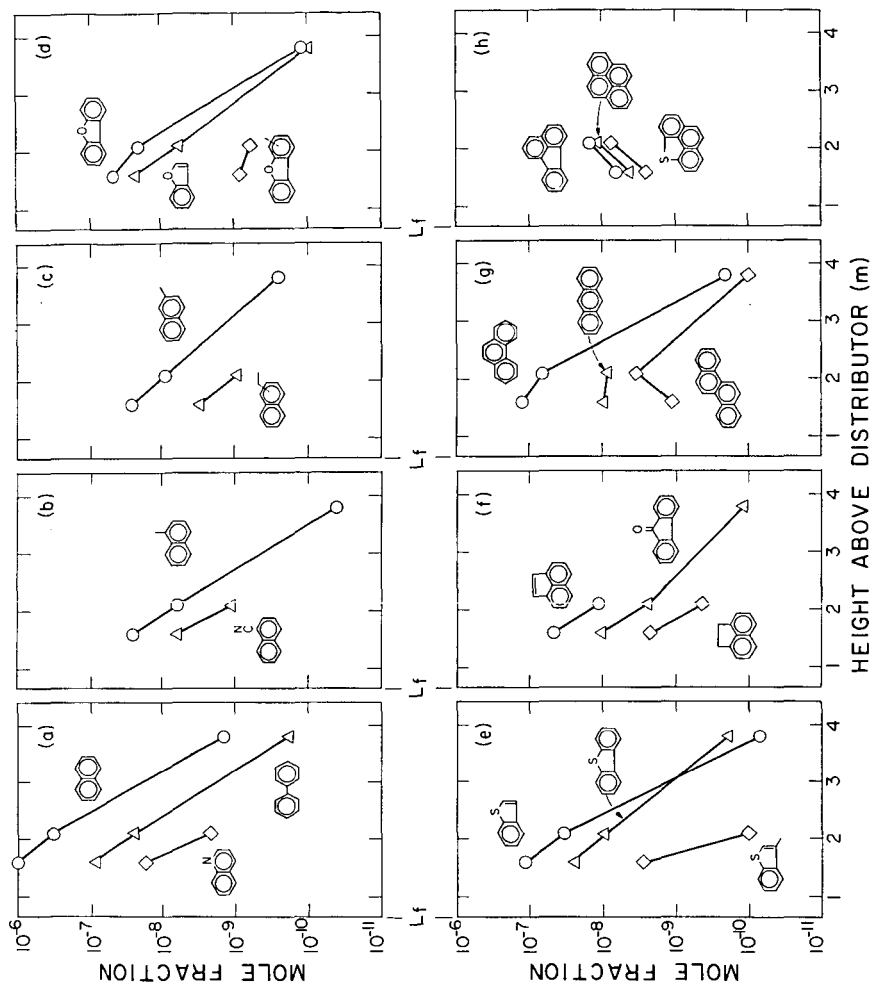


Figure 2. Mole fractions of polycyclic aromatic compounds (dry basis) vs. height in the freeboard.

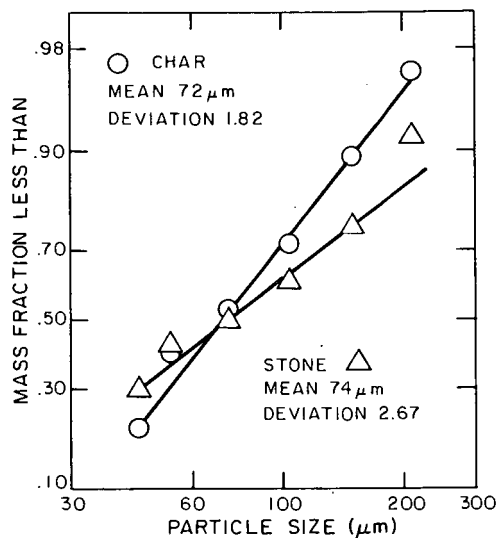


Figure 3. Size distributions of elutriated char and stone.

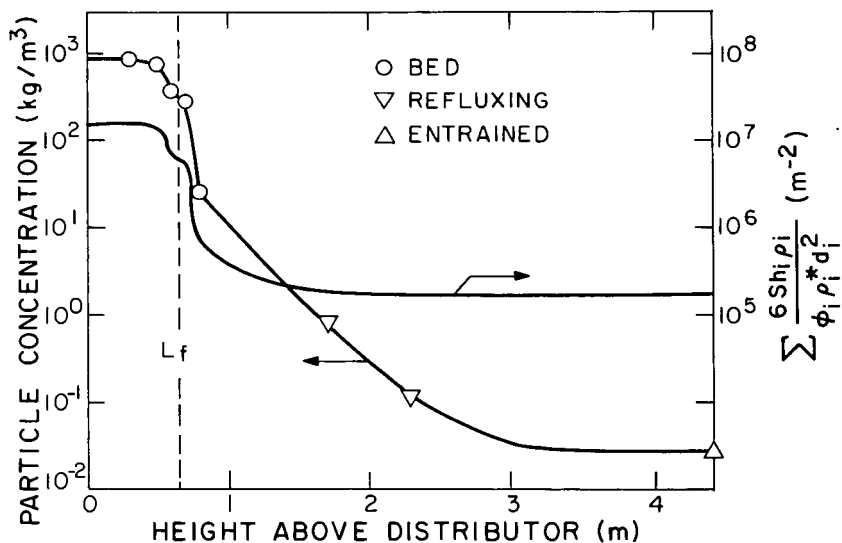


Figure 4. Particle concentration vs. height in the bed and freeboard.

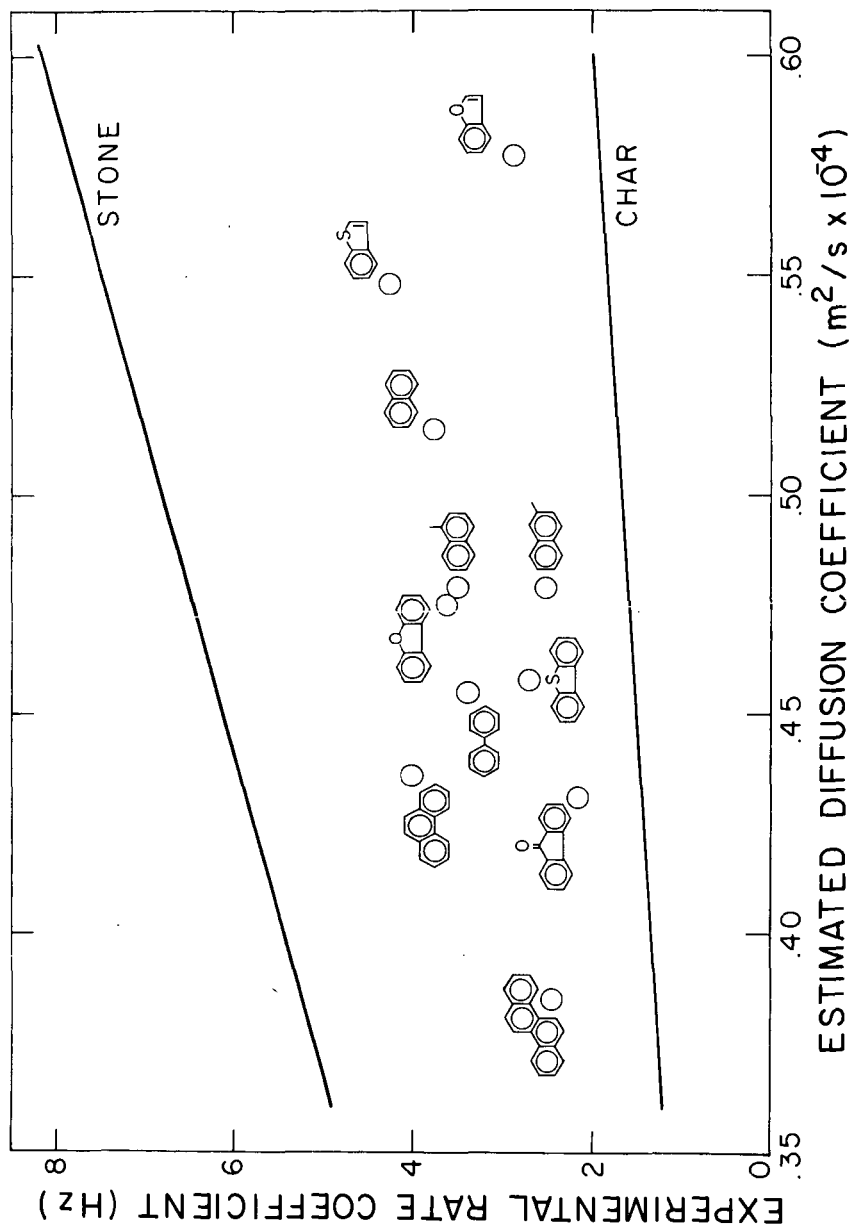


Figure 5. Experimental pseudo-first order rate coefficients vs. estimated molecular diffusion coefficients of the polycyclic aromatic compounds.